

(12)

EUROPEAN PATENT APPLICATION

(21) Application number: 82105092.9

(51) Int. Cl.³: **H 01 M 8/06**
H 01 M 8/04

(22) Date of filing: 11.06.82

(30) Priority: 12.06.81 US 272947

(43) Date of publication of application:
22.12.82 Bulletin 82/51

(84) Designated Contracting States:
DE FR GB IT

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(54) Electrochemical cell system with internal reforming.

(57) A fuel cell system (1) provided with a first passage means (11) in communication with and adjacent to a cell diffusion electrode (2) and with a second passage means (9) means having a catalyst (13) for reforming hydrocarbons and communicating with the first passage means through a gas porous member (8). Differential pressure means is further provided to establish a pressure differential between the passage means (9, 11) for promoting reformed gas flow from the second passage means (9) to the first passage means (11) and retarding electrolyte vapor passage from the first passage means (11) to the second passage means (9).

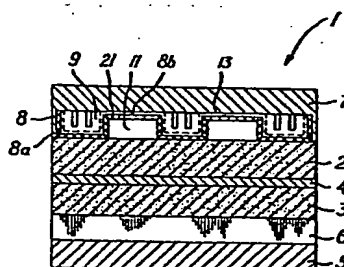


FIG. 1

ELECTROCHEMICAL CELL SYSTEM WITH
INTERNAL REFORMING

Background of the Invention

This invention pertains to fuel cells and, in particular, to fuel cells in which there is internal reforming of the hydrocarbon content of the fuel cell supply gas.

It is known in the design of fuel cells, such as, for example, molten carbonate and phosphoric acid cells, to internally reform the hydrocarbon content of the fuel supply gas. Such hydrocarbon content usually contains methane which itself is relatively electrochemically inactive, but which when reformed produces hydrogen and carbon monoxide which are significantly more electrochemically active and, therefore, can readily participate in the fuel cell reaction. Reforming internal of the fuel cell is beneficial in that the reforming reaction is endothermic and serves to offset heat generated in the cell during operation. Accordingly, by internal reforming, the load on the fuel cell cooling system can be reduced.

U.S. patent 3,488,226 discloses an internal reforming scheme in which the reforming catalyst is situated within the anode electrode gas chamber. The hydrocarbon content of the supply fuel gas is thus reformed during its passage through the anode chamber, and, therefore, is immediately available to the cell anode upon reformation. A drawback of this arrangement, however, is that the endothermic nature of the reforming reaction results in cold spots in the anode chamber which cause condensation of electrolyte vapor transmitted to the anode chamber through

1 the gas-diffusion anode electrode. Such condensation, in
turn, may severely reduce catalytic activity and, as a result,
the reforming reaction.

5 U.S. patent 4,182,895, assigned to the same
assignee hereof, attempts to avoid electrolyte vapor conden-
sation by providing an electrolyte-isolated chamber in which
the catalyst is placed and in which the reforming reaction
takes place. Fuel supply gas reformed in the electrolyte-
isolated chamber is then introduced into the anode (electro-
10 lyte-communicative) chamber for electrochemical reaction.
Owing to the isolation of the reforming chamber from the
electrolyte, electrolyte vapor condensation on the reforming
catalyst does not occur and catalyst activity is preserved.
In this arrangement, reformed gas is not immediately avail-
15 able to the anode chamber, but must be introduced into such
chamber subsequent to reformation.

It is an object of the present invention to
provide a fuel cell having improved internal reforming.

20 It is a further object of the present invention to
provide a fuel cell arrangement in which internal reforming
is carried out such that the reformed gas is made substan-
tially immediately available to the cell electrode, while at
the same time avoiding electrolyte vapor condensation on the
reforming catalyst.

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Summary of the Invention

In accordance with the principles of the present
invention, the above and other objectives are realized in a
fuel cell system wherein a first passage means is provided

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1 adjacent to and in communication with a cell gas-diffusion
electrode and a second passage means having a catalyst for
hydrocarbon reforming communicates with the first passage
means through a gas-porous member. Differential pressure
5 means is further provided to promote flow of hydrocarbons
reformed in the second passage means, through the gas-porous
member and into the first passage means. Reformed gas is
thereby made immediately available to the first passage
means and, therefore, the cell electrode for electrochemical
10 reaction. The differential pressure means, in promoting flow
from the second passage means to the first passage means, also
inhibits electrolyte vapor flow in the opposite direction,
thereby preventing such vapor from reducing catalyst activity
through condensation.

15 In the embodiments of the invention to be disclosed
hereinafter, the gas-porous member includes or incorporates
the reforming catalyst and the differential pressure means
includes constrictions selectively disposed in one or the other
or both of the first or second passage means. Additionally,
20 various catalyst-incorporating gas-porous members are disclosed.

In one form, the gas-porous member comprises a sheet
or plate of solid (substantially non-gas-porous) material which
is made porous by perforating and upon which is disposed a
catalyst layer. In another form, the sheet or plate is made
25 of material which is itself gas-porous and, therefore, need
not be perforated. In yet a further form, the gas-porous
member comprises a gas-porous matrix of a conductive metallic
material into which is impregnated the catalyst material.

1 This form of matrix is advantageous in itself, since it
facilitates conductive contact with the cell electrode.

Brief Description of the Drawings

5 The above and other features and aspects of the
present invention will become more apparent upon reading the
following detailed description in conjunction with the
accompanying drawings, in which:

FIG. 1 shows schematically a fuel cell system in
accordance with the principles of the present invention;

10 FIG. 2 shows an isometric view of the gas-porous
member of the cell of FIG. 1;

FIG. 3 illustrates schematically two adjacent
channels of the gas-porous member of FIG. 2;

15 FIG. 4 shows schematically the pressure differential
along the length of the adjacent channels of FIG. 3.

FIG. 5 shows adjacent channels of a gas-porous
member in accordance with a second embodiment of the present
invention; and

20 FIG. 6 illustrates a gas-porous member of the type
shown in FIG. 5 shaped as a corrugated element.

Detailed Description

25 In FIG. 1, a fuel cell 1 comprises anode and
cathode electrodes 2 and 3, of gas diffusion type, having an
electrolyte matrix 4, therebetween. A separator plate 5
defines a chamber 6 for receiving cathode supply gas and
delivering same to the cathode electrode 3. A further
separator plate 7 in cooperation with valley regions 8a of a
corrugated member 8 defines chambers or channels 9 for
receiving anode supply gas having hydrocarbon content.

30

1 These channels are spaced one from the other by channels 11
defined by the crest regions 8b of the member 8 and the
anode electrode 2.

5 In accordance with the invention, the cell 1 is
further adapted to reform the hydrocarbon content of the
anode supply gas in channels 9 and to deliver such reformed
gas to the anode electrode 2 upon being reformed and in a
manner which inhibits electrolyte deactivation of the
reforming catalyst. More particularly, this is realized in
10 the fuel cell 1 by forming the member 8 as a gas-porous
member incorporating a reforming catalyst and by providing
means for establishing a pressure differential between the
channels 9 and 11 which is positive in the direction of the
channels 11 (i.e. the channels 9 are at higher pressure than
15 the channels 11) so as to promote gas flow from the former to
the latter channels.

In FIG. 1, the gas-porous member 8 is in the form
of a plate of solid material which has been made gas-porous
by placing perforations 21 therein and which supports a
20 porous catalytic layer 13 on its upper surface, i.e., on its
surface bordering the channels 9. Constrictions for selec-
tively constricting flow in the channels 9 and 11 provide the
desired positive differential pressure promotive of gas flow
from the channels 9 to the channels 11 through the gas-porous
25 member 8. FIG. 2 more clearly shows constrictions in the
form of walls 14 disposed at the exit ends 9b of the channels
9 to inhibit gas exit, the input ends 9a of the channels
being open to permit gas entry. As shown in FIG. 2, the
walls 14 have slots 14a so as to only partially constrict the
30

1 respective passages 9 and thereby allow for removal of
undesired reforming reaction products. Further constrictions
in the form of walls 15 are disposed at the entry ends 11a
of the channels 11 to retard gas entry, while the output or
5 exit ends 11b of these channels are, in turn, open to enable
exit of the reaction products of the electrochemical reaction.
The constrictions 15 may also be slotted as at 15a so as to
allow for partial entry of gas, if desired.

10 With the constrictions 14 and 15 properly adjusted,
so as to result in a pressure profile p1, (see FIG. 4) along
the length of the channels 9 which is higher than the
corresponding pressure profile p2 along the length of the
channels 11, as above-described, the anode supply gas
introduced in the input ends 9a of the channels 9, as it
15 passes down the channels, will have its hydrocarbon content
reformed by the catalytic layer 13 (in the case of methane,
hydrogen and carbon monoxide are generated) and the reformed
gas will be urged to pass through the layer 13 and the
perforated plate member 8 into the channels 11. Reformed
20 gas will thus be immediately available to the anode electrode
2 via channels 11 upon being generated.

25 The presence of the positive pressure differential
between the channels 9 and 11 also tends to retard vaporized
electrolyte in the chambers 11 from further migrating into
and through the plate 8 and into the chambers 9. Such
vaporized electrolyte is thus substantially kept away from
the catalytic layer and deactivation of same is avoided.

30 The perforated plate of gas-porous member 8 might
typically be fabricated from a corrosion resistant material,

1 such as, for example, stainless steel, this material then
being provided with fine perforations which might, for
example, be about 50-1000um. The catalyst layer 13, in turn,
5 should preferably be such that only the fine pores of the
member 8 are continuous and may comprise, for example, Ni,
Ni-Cr, Ni-Co, Ni-Mo or suitable combinations of these
materials. The use of the additive materials (Cr, Co, Mo) is
advantageous where a high initial surface area and a high
10 stability for the layer 13 is desired. The Co and Mo addi-
tives, furthermore, also may be helpful in providing H_2 S
tolerance to performance decay, while the Co additive might
additionally enable reforming at low steam-to-carbon (S/C)
ratios.

15 In a modified construction of the porous member 8
of FIG. 1, the member is again in the form of a plate or
foil, but in this case the plate material is itself porous
to the reformed gas generated by the catalytic layer 13.
Thus, for example, where the reformed gas is hydrogen, the
plate might comprise nickel or a nickel alloy or palladium,
20 both of which materials (nickel and palladium), allow diffusion
of molecular hydrogen. The use of such gas-porous materials are
further advantageous in cells wherein the electrolyte matrix 4
comprises molten carbonate as the electrolyte. In particular,
these materials do not react with molten carbonate and, there-
25 fore, do not contribute to electrolyte loss.

Another modification of the porous member 8 of
FIG. 1 is to further provide a thin layer of palladium on
the bottom surface of the plate facing the anode electrode
2. This type of layer will act as a selective membrane to
30 permit reformed gas diffusion through the member 8, while at

1 the same time inhibiting diffusion of large amounts of
water. As a result water will not transfer from the channels
9 to the channels 11, thereby tending to promote the reforming
reaction and to avoid retarding of the electrochemical
5 reaction.

With the fuel cell 1 of FIG. 1 constructed as
above-described, the selective transport of the reformed
anode gas into the channels 11 provides higher efficiency
and improved current density because of the higher reformed
10 gas partial pressures. An overall improved cell thereby
results.

FIGS. 5 and 6 show a second embodiment of a gas-
porous member 8 in accordance with the present invention.
In this case, the member 8 is in the form of a conductive porous
15 metallic matrix whose pores have been impregnated and are filled
with a catalyst material. Such a matrix can be corrugated as
shown in FIG. 6 and used in the same manner as illustrated in
FIG. 1 to define the channels 9 and 11. Alternately, this form
of member 8 can be made into thick rectangular sheets which can
20 then be supported between the plate 7 and the anode electrode 2,
as schematically depicted in FIG. 5. Useable metallic materials
for the matrix are nickel, palladium and other conducting
stable metal alloys acceptable for the fuel cell environment.

The aforesaid conductive matrix form of the
25 porous member 8 is further advantageous, since the conductive
nature of the member promotes coupling of the electrical
energy from the anode electrode through the separator plate
7 to the cell output.

It should be noted that if a more uniform distribution
30 of gas flow into the chambers 11 along their length is desired,

1 the porosity of the gas-porous member 8 can be tailored to
provide the desired uniformity. In general, decreasing the
porosity of the member adjacent the gas entry end (i.e., ends
11a and 9a in FIG. 1) relative to the gas exit end (i.e., the
5 ends 11b and 9b in FIG. 1), will promote increased uniformity in
gas flow distribution. In the particular case of the perforated
plate embodiment of the member 8, this can be accomplished,
for example, by utilizing an increased number of perforations
and/or perforations of increased size at the gas exit end
10 relative to the gas entry end.

In all cases, it is understood that the above-
described arrangements are merely illustrative of the many
possible specific embodiments which represent applications
of the present invention. Numerous and varied other arrange-
15 ments can readily be devised without departing from the
spirit and scope of the invention. Thus, for example, the
catalyst layer 13 need not be incorporated into the porous
member 8, but instead could be placed elsewhere in the
channels 9 such as, for example, on the separator plate 7.
20 Furthermore, the catalyst may be in the form of a pellet bed.
Another modification would be to utilize a catalyst layer 13 on
both the upper and lower surfaces of the plate member 8. Also,
the anode member 2 may include additional components such as,
for example, an electrolyte storage element and/or a current
25 collector, interposed between the anode body and the member 8.

1 What We Claim Is: .

1. An electrochemical cell system comprising:
a gas diffusion electrode;
first passage means in communication with said
5 electrode;
second passage means having a catalyst for
reforming hydrocarbons and including a gas porous member
in communication with said first passage means;
and means for establishing a differential
10 pressure between said first and second passage means so as
to promote gas flow from said second passage means through
said gas-porous member to said first passage means,
whereby the hydrocarbon content of supply gas introduced
into said second passage means is reformed by said
15 catalyst and said reformed hydrocarbon content is aided
by said differential pressure to pass through said gas-
porous member into said first passage means to said gas-
diffusion electrode for electrochemical reaction, and
electrolyte vapor in said first passage means is retarded
20 by said differential pressure from contacting and passing
through said gas-porous member to said second passage
means. .

2. A cell in accordance with claim 1 wherein:
said gas-porous member includes said catalyst;
25 said gas-porous member comprises either a
perforated plate of nongas-porous material or a plate of
gas-porous material;
and said catalyst is a layer disposed on said
plate.

30 3. A cell in accordance with claim 2 wherein:
said layer is on the surface of said plate
facing said second passage means.

4. A cell in accordance with claim 3 further
comprising:
35 a further layer disposed on the surface of said
plate facing said first passage means, said further layer

1 comprising a material allowing selective passage of
products of said reformed hydrocarbon content.

5 5. A cell in accordance with claim 2 wherein:
said layer is on the surfaces of said plate
facing said first and second passage means.

6. A cell in accordance with claim 1 wherein:
said first passage means has first entry and
exit ends:

10 and said second passage means has second entry
and exit ends, said gas-porous member being situated
between said second entry and exit ends.

15 7. A cell in accordance with claim 6 wherein:
said differential pressure means maintains said
differential pressure over the lengths of said first and
second passage means commensurate with the length of said
gas-porous member.

20 8. A cell in accordance with claim 6 wherein:
said differential pressure means includes a
first constriction means selectively disposed in one of
said first and second passage means for constricting the
flow therein.

9. A cell in accordance with claim 8 wherein:
said one passage means is said second passage
means;

25 and said first constriction means includes a
first constriction member disposed adjacent the exit end
of said one passage means for selectively constricting
said exit end of said one passage means.

30 10. A cell in accordance with claim 8 wherein:
said one passage means is said first passage
means;

35 and said first constriction means includes a
first constriction member disposed adjacent the entry end
of said one passage means for selectively constricting
said entry end of said one passage means.

1 11. A cell in accordance with claim 9 or 10
wherein:

 said first constriction member totally constricts
the entry end of said one passage means.

5 12. A cell in accordance with claim 9 or 10
wherein:

 said first constriction member partially
constricts said adjacent end of said one passage means.

10 13. A cell in accordance with claim 8 wherein:
 said differential pressure means further
includes a second constriction means selectively disposed
in the other of said first and second passage means for
constricting the flow therein.

15 14. A cell in accordance with claim 2 wherein:
 said gas-porous member is corrugated so as to
have crest regions and valley regions, said crest regions
serving to partially define said first passage means and
said valley regions serving to partially define said
20 second passage means; and said anode electrode contacts
the peaks of said valley regions to further define said
first passage means; and wherein the cell further
comprises a separator plate disposed in contact with the
peaks of said crest regions to further define said second
passage means.

25 15. A cell in accordance with claim 2 wherein:
 said gas-porous member comprises a conductive
gas-porous matrix;
 and said catalyst is contained in the pores of
said matrix.

30 16. A cell in accordance with claim 2 further
comprising:
 an electrolyte adjacent said gas diffusion
electrode; said gas diffusion electrode being an anode
electrode.

35 17. A cell in accordance with claim 1 wherein:
 the porosity of said gas-porous member is

1 selected to promote uniformity in the flow distribution
of said reformed hydrocarbon content passing into said
first passage means.

5 18. An electrochemical cell system comprising:
a gas diffusion electrode;

a gas-porous conductive matrix at least partially
in contact with said electrode;

a catalyst for reforming hydrocarbons, said
catalyst being disposed in the pores of said matrix;

10 and a conductive plate at least partially in
contact with said matrix and defining a chamber for supply
gas having a hydrocarbon content.

15 19. A cell in accordance with claim 18 wherein:
said conductive matrix is metallic.

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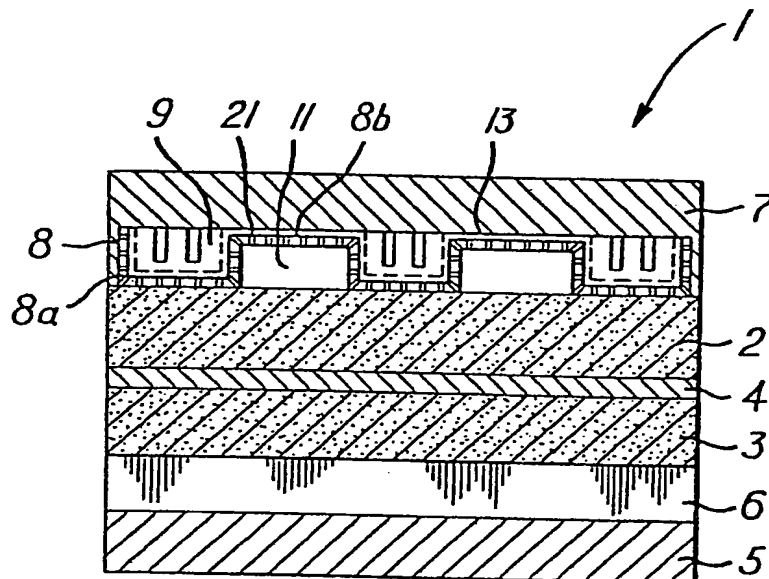


FIG. 1

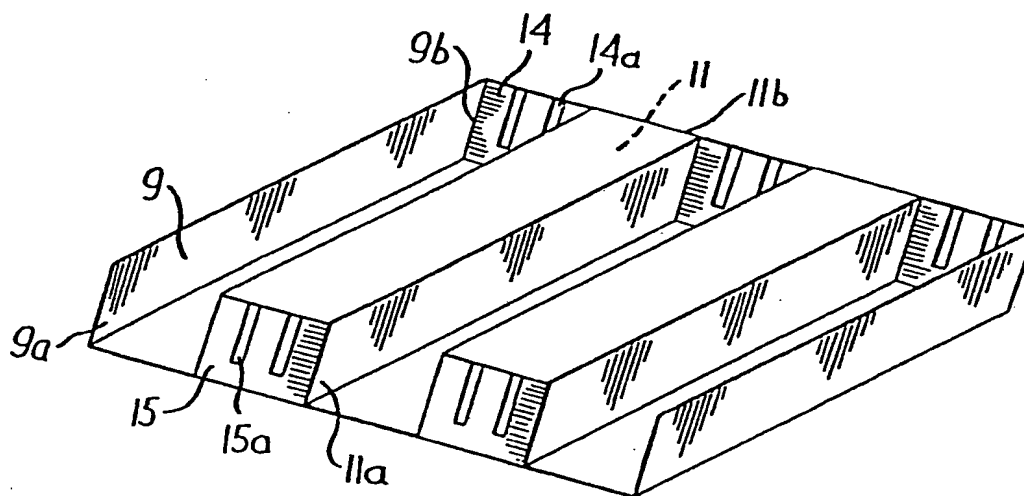


FIG. 2

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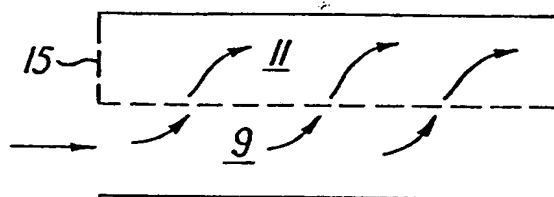


FIG. 3

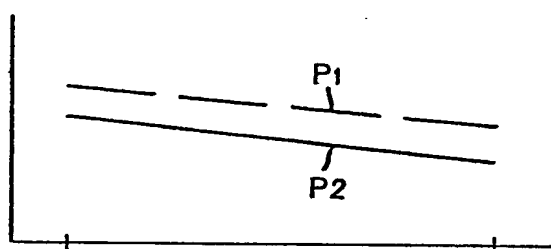


FIG. 4

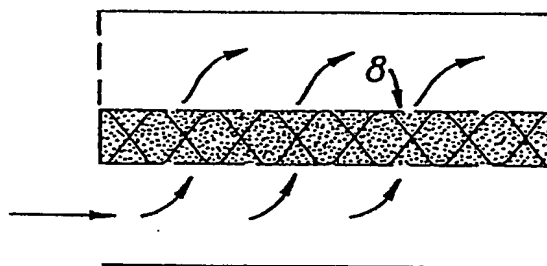


FIG. 5

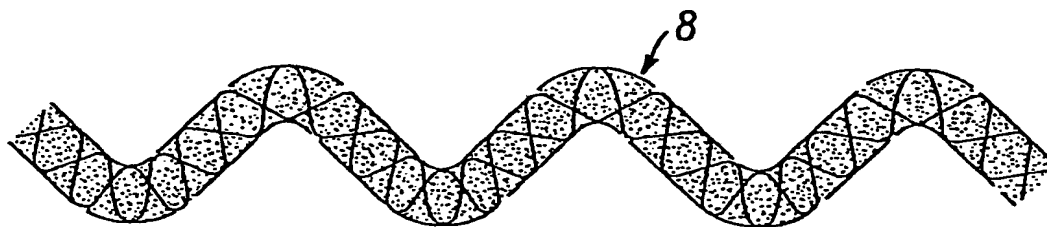


FIG. 6



European Patent
Office

EUROPEAN SEARCH REPORT

0067423

Application number

EP 82105092.9

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. Cl. 3)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
D,A	US - A - 4 182 795 (BAKER et al.) * Column 3, lines 28-60 * --	1	H 01 M 8/06 H 01 M 8/04
D,A	US - A - 3 488 226 (BAKER et al.) * Abstract * --	1	
A	US - A - 4 169 917 (BAKER et al.) * Column 2, line 62 - column 3, line 17 * --	1	
A	US - A - 4 192 906 (MARU) * Abstract * ----	1	TECHNICAL FIELDS SEARCHED (Int. Cl. 3) H 01 M
			CATEGORY OF CITED DOCUMENTS
			X: particularly relevant if taken alone Y: particularly relevant if combined with another document of the same category A: technological background O: non-written disclosure P: intermediate document T: theory or principle underlying the invention E: earlier patent document, but published on, or after the filing date D: document cited in the application L: document cited for other reasons
X	The present search report has been drawn up for all claims		&: member of the same patent family, corresponding document
Place of search VIENNA		Date of completion of the search 16-09-1982	Examiner LUX